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CONVERSION OF URANIUM DIOXIDE TO URANIUM CARBIDE IN AN INDUCTION PLASMA TORCH REACTOR

by

D. Ramaswami, J. Pavlik, and A. A. Jonke

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ABSTRACT

Exploratory studies were conducted to determine the feasibility of using an induction plasma torch reactor for converting uranium dioxide-graphite powder agglomerates to uranium carbide spheres. The agglomerates were fed continuously into the tail flame of a 25-kW (4-MHz) induction plasma torch pointed downward. The solid particles were entrained by the tail flame and were directed into a product receiver, where they were separated from the gas stream.

About 20% of the particles in the product were shiny (with metallic luster) black spheres of 25-50 μm. The remainder of the particles had shapes similar to those of feed particles, with some rounding off of sharp corners.

Between 74 and 82% of the carbon in the feed agglomerates was converted to gaseous and solid products (CO, UC, and alpha-UC_2). The percent carbon utilization was almost constant, although the C/U atom ratio in the feed was varied between 1.4 and 3.2, the average particle size of the feed between 58 and 107 μm, and the solid feed rate between 0.06 and 0.9 kg/hr.

The results of this exploratory study indicate that the method might be suitable for continuous large-scale production, under critically-safe conditions, of (U,Pu)C fuel for fast breeder reactors.

I. SUMMARY

The use of an induction plasma torch reactor for converting $(U,Pu)O_2$ to reactor-grade carbide spheres in high yield was explored. A 25-kW (4-MHz) induction plasma torch was used to convert UO_2 and carbon to UC under various operating conditions, to examine the effects of variables on product characteristics, and to provide a basis for process evaluation. Although the experimental work was devoted to the conversion of uranium dioxide-graphite powder agglomerates to uranium monocarbide, the process is expected to be applicable to the production of $(U,Pu)C$ in equipment installed in an alpha-tight enclosure.

The method involves making composite particles of uranium dioxide and graphite and reacting the composite particles by passing them through and cocurrently with the tail flame (pointed downward) of hydrogen-argon plasma in a 1.5-in.-ID quartz tube. Back reaction is prevented by rapid quenching of the products and by rapid separation of the gaseous product from the solid. The product, in the form of small, free-flowing spheres of carbide, is collected along with unreacted oxide-graphite agglomerates in a product receiver at the bottom of the reactor. The CO reaction product leaves the reaction chamber with the inert plasma gas.

About 20% of the particles in the product were shiny (with metallic luster) black spheres of 25-50 μm. The shapes of the remainder of the particles were similar to those of the feed particles, except for some rounding off of sharp corners. The average particle size for the product was lower than that for the feed because of (1) reaction and spheroidization in the formation of the carbide spheres, (2) densification accompanying sintering of the particles, and (3) breakage of particles due to rapid evolution of the gaseous product (CO).

Between 74 and 82% of the carbon in the feed agglomerates was converted to gaseous and solid products (CO, UC, and alpha-UC_2) in a single pass through the torch in runs with argon in the reaction chamber. These values for the carbon utilization are satisfactory for the 25-kW induction plasma torch used. Although reaction of the graphite was incomplete in these runs, the oxide could probably be completely converted to carbide by passing the solid product from a torch through a second torch or, if only one torch was available, by recycling the solid product.

The carbon utilization was almost constant, although the solid feed conditions were varied--i.e., the solid feed rate was varied between 0.06 and 0.9 kg/hr, the average particle size between 58 and 107 μm, and the C/U atom ratio in the feed between 1.4 and 3.2. Similarly, in a preliminary series of runs with granular alumina, the percentage of particles converted to spheres was almost constant, although the powder feed rate was varied between 0.6 and 1.2 kg/hr. Apparently, the upper limit for the solid feed

rate (where the solids start cooling the plasma flame) has not been reached in the current runs and feed rates much higher than those tested may be practical. Assuming that the potentialities for higher solid feed rates and more efficient operation achieved through the use of metal-walled plasma containment tube compensate for the number of passes required for complete conversion, carbide production rates of at least 0.12 lb/kWh may be expected.

In the conversion of agglomerates to product, carbon and oxygen concentrations decreased in the same proportion; i.e., the value of the ratio, decrease in g-atoms of carbon per g-atom of uranium/decrease in g-atoms of oxygen per g-atom of uranium, was ~1.0; this indicates that the gaseous product of the UO_2-graphite reaction in an argon-3.2 vol % hydrogen plasma flame is CO. X-ray diffraction analysis of the solid-product samples indicated that uranium monocarbide and alpha-uranium dicarbide were present along with unreacted uranium dioxide. The concentration of dicarbide might be lowered to the desired level by a purification step, such as reduction in a hydrogen atmosphere or reaction with uranium dioxide, to form uranium monocarbide.

The current exploratory study indicates that the induction plasma torch method is worthy of further consideration for development to a full-scale facility that is critically safe for continuous conversion of $(U,Pu)O_2$ to carbide spheres. However, the performance of equipment components during the 38 runs with the 25-kW induction plasma torch indicated that the designs of the plasma containment tube, the solids feed orifices, the solids guide tube, and other components would have to be improved for further development or scale-up of the conversion process.

II. INTRODUCTION

Conversion of fast-breeder-reactor oxide to reactor-grade carbide fuel in an induction plasma torch reactor has the potential for converting the oxide-graphite particles to carbide spheres in critically safe equipment that can be continuously operated. Criticality safety and continuous operation are desirable features for commercial production of nuclear fuels. However, the feasibility of the production of carbide spheres from oxide in an induction plasma torch reactor has not been reported in the literature.

The objective of the current study was to determine the feasibility of the production of the carbide spheres using an induction plasma torch reactor. The experimental work was limited to the conversion of uranium dioxide and graphite powder agglomerates to uranium monocarbide, but the process is expected to be applicable to the production of (U,Pu)C (which would have to be done in equipment installed in an alpha-tight enclosure). The objective of the experimental work was to determine the operating conditions required to produce high yields of reactor-grade uranium monocarbide.

A schematic flowsheet for the conversion of $(U,Pu)O_2$ to $(U,Pu)C$ spheres using an induction plasma torch reactor is shown in Fig. 1. The method includes (1) preparing 100-μm composite particles of $(U,Pu)O_2$ and carbon, (2) reacting the composites by passing them through and cocurrently with the plasma flame of an inductively coupled plasma torch pointed downward, (3) separating $(U,Pu)C$ product spheres from the nonspheres and unreacted particles, (4) separating the CO reaction product from the plasma gas, and (5) recycling the unreacted particles, nonspheres, and the plasma gas to the reactor.
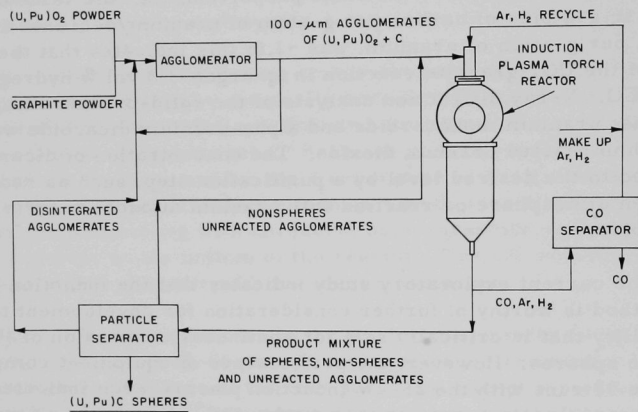
Fig. 1. Conversion of $(U,Pu)O_2$ to Carbide Spheres in an Induction Plasma Torch Reactor

Of the methods described in the literature for preparing composite particles of $(U,Pu)O_2$ and carbon, the better-developed one[1,2] consists of mixing 1- to 2-μm particles of $(U,Pu)O_2$ and graphite in a V-blender together with a small amount of binder such as polyvinyl alcohol. After the dry powder is mixed, a small amount of water is added and mixing is continued until the agglomerates form and finally reach the desired size. The agglomerates are dried at 50°C. Other ways of preparing intimate mixtures of $(U,Pu)O_2$ and carbon are from a nitrate solution containing sugar[3,4] or perhaps from a mixture of the oxides and sugar solution. Any of these methods can be adapted to the preparation of the feed required for the plasma torch method.

In the induction plasma torch reactor, the composite particles of $(U,Pu)O_2$ and graphite are passed through and cocurrently with the plasma flame at high velocities in the form of a tubular column. The residence times of particles in the flame are of the order of a few milliseconds. Particle heating occurs by several mechanisms, including thermal conduction through the boundary layer surrounding the particle, radiation, electron

bombardment, and electron recombination on the particle surface. The extent of conversion of the oxide to the carbide depends on the time/ temperature history of the particles. Back reaction is prevented by rapid quenching of the products and rapid separation of the gaseous product from the solid.

The solid product, in the form of small, free-flowing spheres of carbide accompanied by unreacted oxide-graphite particles, is collected in a product receiver at the bottom of the reactor (although a different experimental setup might provide for continuous transport of the product to the next process step). Commercially available equipment could be used to separate high-density carbide spheres from nonspheres and low-density unreacted agglomerates. Incompletely reacted material would be recirculated to the reactor or to an agglomerator system, depending on particle size. The dense spheres of carbide would be fabricated into fuel elements.

The gaseous reaction product, CO, leaves the reaction chamber with the inert plasma gas, argon (or hydrogen-argon mixture). The carbon monoxide is separated from the plasma gas by well-known commercial methods,[5-7] and the plasma gas is recycled to the torch.

III. LITERATURE REVIEW

Spheroidization of various powders using induction plasma torches has been reported in the literature, as well as synthesis and spheroidization of uranium carbides with dc-arc plasma jets. Work related to the expected performance of an induction plasma torch reactor for the conversion of uranium dioxide-graphite agglomerates to uranium carbide spheres is reviewed here.

A. Spheroidization Studies Using Induction Plasma Torches

An induction plasma torch (10 kW, 5 MHz) enclosed in an alpha box was used by Jones et al.[8,9] for producing thermally and chemically stable microspheres of plutonium dioxide. The microspheres were 10-250 μm in diameter, the majority were dense, solid, crystalline particulates, and a small percentage had internal voids. The density of the product was 96% of the theoretical crystalline density.

Browne and Latta[10] reported spheroidization of UO_2, ThO_2, and ZrO_2 particles with a 7.5-kW (5-MHz) induction plasma torch.

Hedger and Hall[11] studied spheroidization of the powders of chromium, molybdenum, tantalum, tungsten, alumina, magnesia, and several compounds of uranium using a 25-kW induction plasma torch. The studies were mainly

confined to 100- to 150-μm particles, and powder feed rates of 0.06 to 0.3 kg/hr were used. Yields of 50 to 70% spheroidization (in a single pass through a plasma torch) were obtained.

Tremper[12] used a 10-kW induction plasma torch for the spheroidization of ceramic powders. He found that the frequency range of 4-6 MHz was best for spheroidization, almost entirely because in this range plasma was the most stable. For most of the work, the high-frequency power was furnished through a four-turn cylindrical coil (made of 3/16-in. copper tubing) at a frequency of 4 MHz. A solids feed nozzle that gave laminar flow of the powder provided a much higher degree of spheroidization than any other nozzle tried by him. Powder feed rate was 0.06 kg/hr. For a single pass of 43- to 61-μm silica powder through the plasma torch, the powder was approximately 75% spheroidized; two passes of the powder through the torch gave about 90% spheroidization. The resulting silica spheres were all nearly 100% dense, with only a few small bubbles in them. The spheroidization of 40-μm alumina upon one pass through the induction plasma torch was about 65%, upon two passes 80%, and upon three passes 90%. About half of the alumina spheres produced had nearly theoretical density.

B. Synthesis and Spheroidization of Uranium Carbides

A dc-arc plasma jet method of making spherical actinide carbide was patented by White and O'Rourke.[13] In this method, agglomerates (70 to 150 μm) of UO_2 and carbon fed to a dc-arc plasma are entrained by the jet. The actinide carbide product flowing from the tip of the jet consists of completely reacted, spherical (50 to 125 μm) particles. A small amount of hydrogen, less than 1 vol %, added to the inert plasma gas, acted as a promoter that helped drive to completion the reaction of the UO_2 and carbon.

Gibson and Weidman[14] used a consumable homogeneous anode of UO_2 and carbon to produce a uranium-carbon plasma. The temperature at the tip of the anode is raised to 3700-4700°C in a chamber at a pressure of 0.1-5 mm Hg. The very high temperature initiates the extremely rapid reaction

$$UO_2(s) + 3C(s) \rightarrow UC(s) + 2CO(g),$$
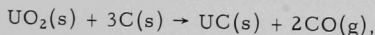
and the solid product formed quickly melts. The molten uranium carbide on the anode face becomes a zone that is heated to the boiling point of the system. The liquid product that drips from the reaction zone is completely outgassed of lower-boiling-point impurities, and a high-density, high-purity solid product is collected. The density of uranium carbide spheres produced in this manner was 97 to 99+% of theoretical, the purity was 99.95%, and the diameters were 44 to 6000 μm. The production rate was about 0.18 lb of UC per kWh of dc input.

Synthesis of uranium carbide by the gas-phase reaction of UF_6 with methane and hydrogen was investigated by Helton,[15] who used a dc-arc plasma jet. Some uranium carbide was formed when finely divided SiC was premixed with the gaseous mixture and then introduced into the plasma jet. Analysis of the product by X-ray diffraction showed that small quantities of uranium dicarbide and uranium monocarbide were present. However, yields were never greater than 1%.

Synthesis of uranium carbide from a mixture of uranium tetrafluoride and silicon carbide was explored by Helton,[15] who used an induction plasma torch. A mixture of powdered uranium tetrafluoride and silicon carbide was fed to the torch at a rate of 0.06-0.12 kg/hr. The solids (product) obtained from the reaction were analyzed by X-ray diffraction. A trace of uranium monocarbide was present. Quantitative yields were never greater than ~1%.

Jets of plasma generated by passing a gas through a dc arc were extensively used in early investigations on spheroidization of carbide particles. Sturge and Smyth[16,17] investigated the spheroidization of $(U,Th)C_2$ with a 20-kW dc-arc argon plasma jet. For 500-μm agglomerates fed at 0.2 kg/hr, spheroidization was better than 99%. Throughputs greater than 0.2 kg/hr tended to produce ovoids. For 250- to 353-μm agglomerates of UZrC with 24-kW input dc, spheroidization rates of 0.2 kg/hr were reported by Jacques and Sturge.[18] Bildstein[19] reported an investigation using a dc-arc plasma jet in which $(U,Th)C_2$ and UC_2 particles up to 500 μm, and UZrC particles up to 350 μm, were spheroidized in one pass to the extent of 98%.

C. Expected Performance of Plasma Torch

From the above literature review, factors affecting the conversion of uranium dioxide to uranium carbide were inferred assuming that (1) the yields of spheroidized particles described above represent the quantity of heat transferred from a plasma to particles, (2) the reaction kinetics do not limit the extent of reaction, and hence (3) the yields indicate the extent of endothermic reaction to be expected with an induction plasma torch. Conversion of uranium dioxide to uranium carbide was expected to be affected by operating conditions in the following manner:

1. With powder feed rates of 0.06-0.3 kg/hr to a 25-kW induction plasma torch, product yields should be 50-70% in a single pass of 100-μm particles, since in other investigations spheroidization yields were in this range for these operating conditions.

2. The product yield should increase with an increased number of passes through the plasma torch.

3. Yield should increase if a diatomic gas such as hydrogen is added to the plasma gas.

4.   Yields should be higher with a solids feed nozzle that gives laminar flow of the powder than with other types of nozzles.

5.   Product spheres should have near-theoretical density.

6.   Torch operation in an alpha-tight enclosure should be feasible.

IV. EQUIPMENT AND OPERATING PROCEDURE

A. Equipment

The equipment for study of the conversion of uranium dioxide to uranium carbide consists of an induction plasma torch, an rf power supply, a water-cooled reaction chamber, a product receiver assembly, a powder feeder, a gas supply system, and associated instrumentation. A schematic diagram of the plasma-torch installation is shown in Fig. 2.
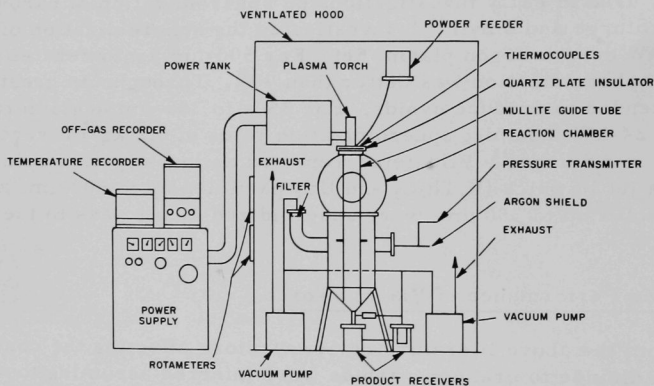
Fig. 2. Installation of Plasma Torch for Studying Conversion
of Uranium Dioxide to Uranium Carbide

The induction plasma torch is held on a quartz plate insulator on the top flange of the water-cooled reaction chamber (see Fig. 3). The plasma torch (TAFA* Model 56) consists of a 1.5-in.-ID quartz tube surrounded by an induction coil immersed in cooling water. About 3 in. of the tube length is covered by the induction coil; a plastic casing enclosing the tube and coils is supplied with cooling water by passages that also carry the rf power leads. The torch is designed so that plasma generation occurs only within the water-cooled quartz tube. The rf coil is energized by a Lepel Model T-25 vacuum-tube high-frequency generator rated for 25 kW.**

*TAFA Division, Humphreys Corporation, Concord, N. H.
**Lepel High Frequency Laboratories, Inc., Queens, N. Y.

The power is delivered to the torch at a frequency of 4 MHz. Additional details on the design of the torch may be found in the manufacturer's literature.[20] The basic principles of rf plasma formation are discussed in detail by Baddour and Timmins.[21]
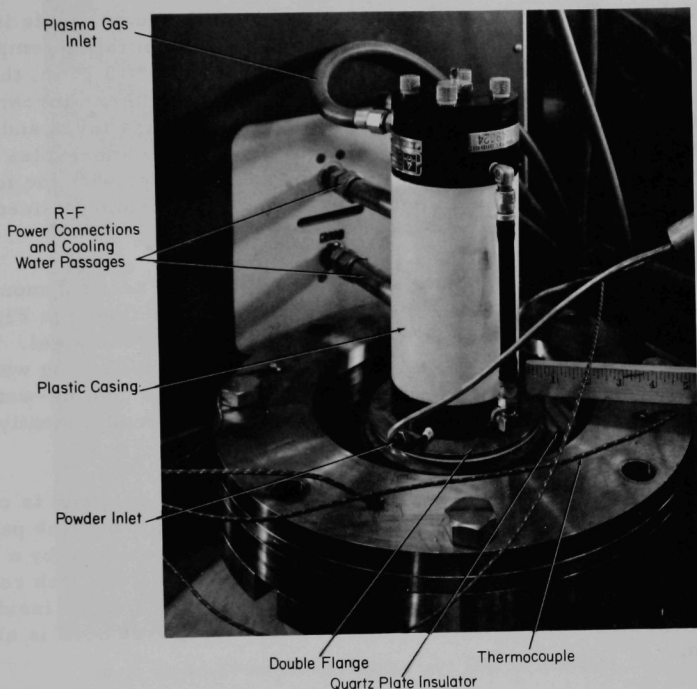
Fig. 3. Plasma Torch Mounted on Top Flange of Reaction Chamber. ANL Neg. No. 308-2115.

The plasma gas, argon (or hydrogen-argon mixture), is fed in a controlled pattern to the quartz tube through inlets located in the gas-injector retainer at the top of the tube. The gas may be introduced radially, tangentially, or axially into the tube to establish the desired flow pattern. Argon and hydrogen are supplied from a cylinder manifold at the desired flow rates. Controls prevent startup of the torch without adequate flow of the gases. The flow rates are metered with rotameters.

The agglomerated uranium dioxide-graphite powder is fed to the plasma torch by a Model 104 power feeder supplied by TAFA. The powder feeder operates on a vibrating feed principle. Up to 0.5 kg of powder is loaded into a bowl, and the vibration intensity of the bowl is controlled with a rheostat. The powder drops into a collecting tube and is transported by

the carrier gas (argon) to the plasma torch. The agglomerates are fed into the flame downstream from the plasma-generation zone. The powder inlet is located in the nozzle at the bottom of the plasma containment (quartz) tube.

A brass double flange holds the torch and the quartz plate insulator that supports the torch. The maximum permissible operating temperature for the brass double flange is 400°C. The quartz plate* (0.75 in. thick with a 4.625 in. ID and a 10 in. OD), in addition to supporting the torch, serves as a thermal and electrical insulator between the plasma torch and the top flange of the water-cooled reaction chamber. Four thermocouples are affixed on the top flange. Temperature records obtained with the four thermocouples are used as an index of thermal equilibrium attained in the reaction chamber.

The reaction chamber is a 28-in.-ID spherical section, mounted on the top of an 18-in.-ID, 40-in.-deep cylindroconical section (see Fig. 2). The entire reaction chamber is made of Type 304 stainless steel. The two sections of the reaction chamber are double-walled with cooling water flowing through the annulus between the two walls. The cooling-water flow rates for the two sections are controlled and metered independently of each other.

A mullite guide tube (5.6 in. ID, 6.1 in. OD, 27 in. long) is centrally located below the torch to contain the tail flame and to direct the particles into the product receiver. The chamber pressure is recorded by a pressure transmitter and recorder assembly. The induction plasma torch reactor assembly is shown in Fig. 4. The reactor assembly is located inside a ventilated hood. The ventilation-air flow rate through the hood is about 1500 cfm.

The product receiver assembly (see Fig. 5) consists of a primary receiver (4 in. ID and 7.5 in. high) for the product, a secondary receiver containing a bayonet-type sintered-nickel filter (1.75 in. wide, 6 in. long), and a bypass line that contains a nickel-wool filter. This type of product receiver assembly is designed to separate the solids from the gas stream while steady flow patterns and pressure are maintained inside the plasma torch. The large particles (the major fraction of the solids) are separated from the gas stream in the primary receiver. The fine particles entrained in the gas stream are retained on the sintered-nickel filter in the secondary receiver. If the pressure in the reaction chamber tends to build up rapidly, the bypass line containing the nickel-wool filter can be opened to exhaust the gases and relieve the pressure. The off-gas (containing the gaseous reaction product, CO) is exhausted from the product receiver assembly by two Duo-Seal vacuum pumps (free-air pumping rates of 800 and 300 ft^3/hr) operating in parallel.

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*Manufactured by the Amersil Corp.; cut, polished, and examined under polarized light by Optical Shops (Central Shops, ANL).
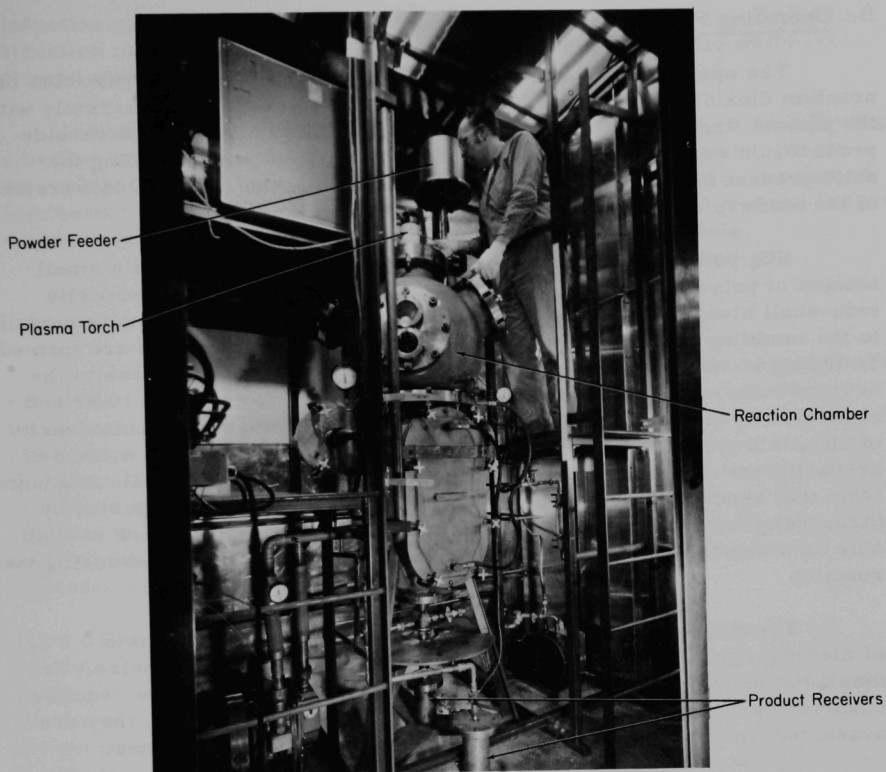
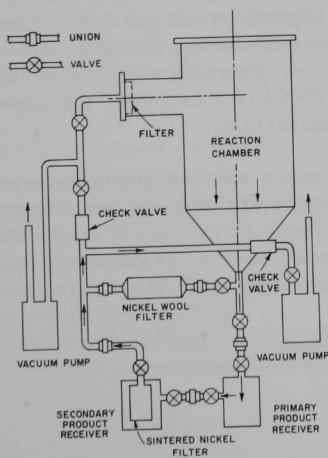Fig. 4. Induction Plasma Torch Reactor. ANL Neg. No. 308-2114.

Fig. 5
Product Receiver Assembly

B. Operating Procedure

The operating procedure includes (1) making composite particles of uranium dioxide and graphite, (2) passing them through and cocurrently with the plasma flame of the induction plasma torch, (3) collecting the carbide product in a receiver at the bottom of the reactor, and (4) sampling the solid product for subsequent analysis to determine the extent of conversion of the oxide to carbide.

UO_2 powder* (0.5-1 μm), graphite powder** (~3 μm), and a small amount of polyvinyl alcohol binder† are dry-mixed in a Patterson-Kelly twin-shell blender for at least 16 hr. Distilled water is added incrementally to the tumbling mixture in the blender until small agglomerates are formed. Tumbling is continued until the size of the agglomerates increases to the desired value. The agglomerates are dried at 50°C for at least 16 hr and separated by sieving. The resulting particles (107 μm) have a bulk density of about 2.8 g/ml and a particle density of about 4.6 g/ml. The method of preparing the composite particles of uranium dioxide and graphite is adapted from that reported by Strausberg.[1] The polyvinyl alcohol is reported by Strausberg to contribute 10% of its initial weight as carbon, which is available as a reactant; the remainder of this compound is volatilized during the reaction.

The powder feeder is loaded with a specified quantity (up to 0.5 kg) of the UO_2-carbon agglomerates of specified stoichiometry and size. To displace the ambient air completely from the powder feeder, the reaction chamber, and the product receiver assembly (see Figs. 4 and 5), they are evacuated (to 70 μm Hg) and filled with argon, cyclically six times.

The desired flow rates for plasma gas and cooling water (to the torch and to the reaction chamber) are established. The rf coil is energized and the input current is increased incrementally until the torch is lit at about 8 kW. The gas flow rates and the input current are then increased to predetermined values. The temperature of the top flange of the water-cooled reaction chamber gradually increases, starting when the torch is lighted, and reaches a steady value (about 85°C) in 1 hr, when the apparatus has reached thermal and electrical equilibrium.

After the induction plasma torch reactor attains electrical and thermal equilibrium, powder feeding is started and continued for the predetermined feed period. Then the torch is turned off by reducing the input current to zero, and the product receivers are isolated. After power is cut off, gas and water flows are continued for a few hours to cool the

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*Supplied by Nuclear Fuel Services, Erwyn, Tennessee.
**Supplied by National Carbon Company, New York, New York.
†Supplied by J. T. Baker & Co., Phillipsburg, N. J.

induction plasma torch reactor. The procedures outlined in the TAFA in-
struction manual for preventing possible hazards are followed while the
plasma torch is being operated.

The isolated product receivers are transferred to a glovebox having
a helium atmosphere to avoid exposure of the solid product to oxygen in
the ambient air and to prevent possible oxidation. The product is sampled
for identification of its components by X-ray diffraction analysis and for the
determination of free carbon, total carbon, and oxygen contents.

V. RESULTS AND DISCUSSION

Thirty-eight runs were made in the current exploratory study on the
conversion of uranium dioxide to uranium carbide in an induction plasma
torch reactor. The initial 21 spheroidization runs (discussed in Appendix A)
were made to determine the operating characteristics of the plasma torch.
Granular alumina was used as the solid feed in the initial 21 runs because
of ease in reactor operation with alumina feed. Agglomerates of UO_2 and
graphite powder were used as the solid feed in the remaining 17 runs. The
detailed operating conditions and data for all 38 runs are tabulated in
Appendixes A and B.

The ranges of operating conditions tested for the conversion of
uranium dioxide-graphite agglomerates to uranium carbide are given in
Table I. These ranges of operating conditions were selected based on the
results of the initial runs. Formation of carbide spheres, utilization of
carbon in chemical reactions, production rates of carbides, identification
of gaseous and solid products, and process scale-up considerations are
discussed below.

TABLE I. Ranges of Conditions Tested for
Conversion of Uranium Dioxide-Graphite Agglomerates
to Uranium Carbide in Induction Plasma Torch Reactor

	Low Value	High Value
C/U atom ratio in the feed	1.4	3.2
Particle size of the feed agglomerates, μm	53	125
Feed rate, kg/hr	0.06	1.2
Feed duration, min	2	60
Power input to the oscillator, kW	15.2	20.0
Plasma gas flow rate, scfh	131	158
Concentration of hydrogen in plasma gas	0	6.0

A. Formation of Carbide Spheres

Some spherical, shiny (with metallic luster) black particles of 25-50 μm were observed in product samples. Typical uranium carbide spheres produced in the induction plasma torch reactor are shown in Fig. 6. These spheres apparently are very dense.
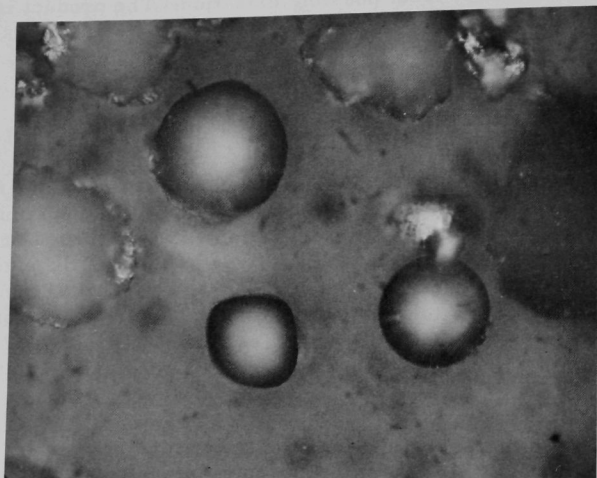
Fig. 6. Typical Uranium Carbide Spheres Produced in
Induction Plasma Torch Reactor. Mag. 500X.

About 20% of the product particles were spheres. The remainder of the particles had shapes similar to those in the feed, with some rounding off of sharp corners. Figure 7 shows the uranium dioxide and graphite agglomerate feed powder and the carbide product of Run 18. The product is typical of that obtained in one pass of the agglomerates through the induction plasma torch. Figure 6 is a magnified view of the spheres at the center of Fig. 7b.

The average particle size was lower for the product than for the feed because (1) reaction and spheroidization had occurred, (2) densification accompanied sintering of the particles, and (3) breakage accompanied the rapid evolution of gaseous product (CO). Methods are commercially available for separating spheres from nonspheres. Hence, in the present exploratory study, no attempt was made to demonstrate the feasibility of separating carbide spheres from nonspheres in the product.

The operating conditions and the characteristics of the induction plasma torch system influence the degree of spheroidization and the percentage of carbide spheres in the solid product. Determination of the effects of these factors on the yield of carbide spheres was beyond the scope of the present study.
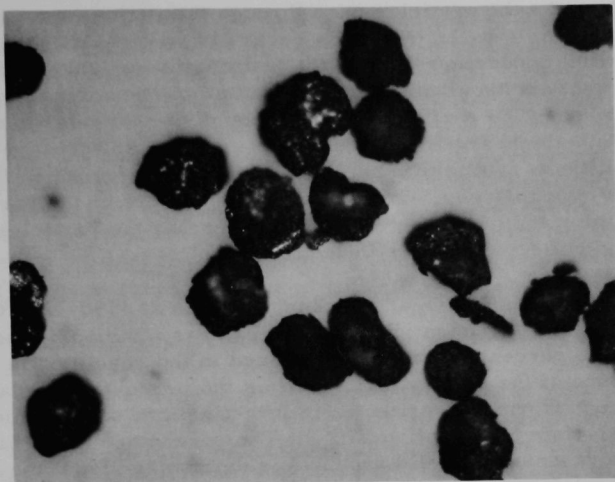
a. Uranium Dioxide and Graphite Agglomerates. Mag. 200X.
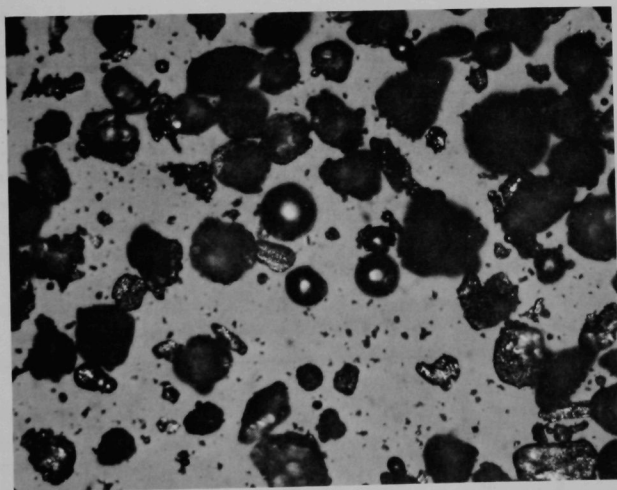
b. Carbide Product after One Pass of the Agglomerates
through the Induction Plasma Torch. Mag. 200X.

Fig. 7. Typical Uranium Dioxide-Graphite Agglomerates and Carbide Product (Run 18)

B. Utilization of Carbon in Chemical Reaction and Production Rates of Carbide

Composite particles of UO_2 and carbon at room temperature are fed to the plasma flame, which is pointed downward inside the induction plasma

torch reactor. As the particles pass through the various temperature zones of the plasma flame, particle heating occurs by several mechanisms, including thermal conduction through the boundary layer surrounding a particle, radiation, electron bombardment, and atom recombination on the particle surface. The particles falling through the hot central zone of the flame reach the highest temperatures, greater than 2500°C (the melting point of UC); those falling through the cooler peripheral zone of the flame reach lower temperatures. The average temperature of the particles increases rapidly, then falls to room temperature as the particles are quenched and collected in the product receiver.

The reactions of UO_2 and carbon[22,23] begin at 1150-1200°C, and the reaction rates are dependent on the temperatures attained by the composite particles. The percentage of carbon utilized in the reactions is the measure of performance of the plasma torch, just as the percentage of particles melted is the performance criterion in investigations of spheroidization of granular materials (see Section III.A.). The percentage of carbon in the feed converted to gaseous and solid products is determined from the total carbon in the feed agglomerates and from the total carbon and free carbon in the solid product thus:

$$\text{Utilization of carbon} = \frac{\left\{\begin{matrix} \text{g atoms of C per g atom of U} \\ \text{in the feed agglomerates} \end{matrix}\right\} - \left\{\begin{matrix} \text{Unreacted g atoms of C per g} \\ \text{atom of U in the solid product} \end{matrix}\right\}}{\left\{\begin{matrix} \text{g atom of C per g atom of U} \\ \text{in the feed agglomerates} \end{matrix}\right\}}.$$

The utilization of carbon was 74-82% (see Table II). These data suggest that at least 74-82% of the particles, which were at room temperature when fed into the plasma flame, might have reached temperatures above 1200°C.

TABLE II. Utilization of Carbon in Chemical Reactions

Plasma gas flow rate: 132 scfh
Power input to oscillator: 15.9 kW
Powder feed: Tail flame of the plasma torch

Run	Composition of Uranium Dioxide-Carbon Agglomerate			C/U Atom Ratio in the Feed	Nominal Feed Rate of Agglomerates, kg/hr	Duration of Feeding, min	Average Particle Size of Agglomerates, μm	Utilization of Carbon, % carbon in feed
	U, wt %	C, wt %	O, wt %					
15	79.2	8.7	12.1	2.2	0.9	10	107	82
16	79.2	8.7	12.1	2.2	0.9	10	107	82
17	81.3	5.7	13.0	1.4	0.9	60	107	80
18	74.7	12.1	13.2	3.2	0.06	60	58	74

The values of carbon utilization achieved in the current work are slightly higher than the percentages of particles melted (50-70%) by making a single pass of granular materials through a 25-kW induction plasma torch at other sites (see Section III.A.). Results for the runs with agglomerates may also be compared with those from the initial runs of the current

work, in which granular alumina was fed into the plasma zone and 89% of the alumina granules melted under operating conditions somewhat similar to conditions for runs with agglomerates. The carbon-containing agglomerates have less favorable emissivity and thermal conductivity compared to alumina granules, and the carbon-containing agglomerates are more difficult to heat than alumina granules. Hence, the value of utilization of carbon reported above appears to be the maximum limit for this system with particle injection into the tail flame of the plasma torch. These results indicate that to attain complete conversion of the oxide to the carbide, either the solid product from the torch has to be passed through a second torch or (if only one torch is available) the product has to be recycled.

The carbon utilization was almost constant (within experimental error) although the solid feed conditions were varied--i.e., the solid feed rate was varied between 0.06 and 0.9 kg/hr, the average particle size between 58 and 107 μm, and the C/U atom ratio in the feed between 1.4 and 3.2 (see Table II). Similarly, in the initial series of runs with alumina, the percentage of particles converted to spheres was almost constant although the powder feed rate was varied between 0.6 and 1.2 kg/hr. Apparently, feed rates for uranium dioxide-graphite agglomerates higher than 0.9 kg/hr or 0.12 lb/kWh would not reduce utilization of the reactants, and the upper limit for feed rates (where the solids start to cool the plasma flame) was not reached in the current runs.

Operating conditions for the current runs included low-enthalpy argon-3.2 vol % hydrogen plasma flame, at low power inputs (15.9 kW) to the oscillator. To operate the torch with high-enthalpy plasma flame, it may be necessary to install a plasma containment tube protected by a metal shield in place of the quartz tube. With this modification, higher overall carbide production rates may be obtained than with the present equipment. Another modification suggested is an improved design of solids-injection orifices to effect higher solids introduction rates than in the current study, which may result in higher carbide production rates. Performance of equipment components is discussed in Appendix C.

On the basis of current results, carbide production rates of at least 0.12 lb/kWh can be expected, if it is assumed that the potential for higher feed rates and more efficient operation related to the use of a metal-walled plasma containment tube is offset by the need for several passes for complete conversion. This production rate can be compared with that (0.18 lb of UC per kWh) achieved by Gibson and Weidman,[14] who used an anode of UO$_2$ and carbon forming a dc-arc plasma.

C. Identification of Gaseous and Solid Products

Information on gaseous and solid products of the reaction between UO$_2$ and carbon at the high temperatures (about 2500°C) attained by particles in the induction plasma torch is useful for process design and

scale-up considerations. The gaseous product was identified from the re-
sults of analyses of the solid product for uranium, carbon, and oxygen.
Solid products were identified by X-ray diffraction analysis.

The ratio of decreases in carbon and oxygen upon conversion of the
feed to product (g-atoms of carbon decrease per g-atom of uranium/g-atoms
of oxygen decrease per g-atom of uranium) was ~1.0 for Runs 15, 16, and 18.
This value for the ratio indicates that the gaseous product of the reaction
between UO_2 and graphite in an argon-3.2 vol % hydrogen plasma flame is CO.

X-ray diffraction analysis of the solid-product samples indicates
that uranium monocarbide and alpha-uranium dicarbide were present, along
with unreacted uranium dioxide (see Table VI later for detailed results).
Possibly, alpha-UC_2 was produced in the hot zone of the plasma flame,
and quenching of the product particles in the reaction chamber prevented
its decomposition to UC and C.[24] The following reactions probably occurred:

$$UO_2 + 3C \rightarrow UC + 2CO; \tag{1}$$

$$UO_2 + 4C \rightarrow alpha-UC_2 + 2CO. \tag{2}$$

Almost identical X-ray diffraction patterns resulted for product
samples from Runs 16 and 18. In these runs, agglomerates with different
carbon contents (8.7 and 12.1 wt % carbon) were fed to the plasma torch at
rates 0.9 and 0.06 kg/hr, respectively. The results suggest that the rela-
tive rates of monocarbide and dicarbide formation are independent of feed
composition, feed rate, and feed particle size (in the range of 58-107 μm).
The extent of conversion of uranium dioxide to uranium carbides appears
to be governed by the temperature level attained by the individual particles.

In a final purification step, the concentration of the uranium di-
carbide may be lowered to the desired level by reducing the hyperstoichio-
metric carbide in a hydrogen atmosphere[25] or by converting the dicarbide
to monocarbide by reaction with uranium dioxide.[26] The presence of large
amounts of the dicarbide along with the monocarbide may be undesirable in
a fuel for a sodium-cooled reactor, because the hyperstoichiometric carbide
is decarburized by the stainless steel cladding, and the fuel-to-cladding
sodium bond facilitates decarburization.[27]

D. Process Scale-up Considerations

In the induction plasma torch method, since the solids and gas are
continuously moving as a dilute suspension at very high velocities, solids
do not build up in any portion of the equipment and large-scale equipment
can be designed to be critically safe.

The design characteristics and operating conditions of the induction plasma torch system influence the conversion of oxide to carbide as well as the percentage of carbide spheres in the solid product; probably, yields of carbide spheres will be higher in large units than in small units. Production rates of at least 0.12 lb/kWh can probably be achieved with more efficient, larger plasma torches than the one used in the present study. A discussion of the economics of plasma torch operation by Dundas and Thorpe[28] indicates that a large-scale unit may possibly be operated economically.

APPENDIX A

Preliminary Runs Using Granular Alumina

In a series of preliminary runs, granular alumina (44-149 μm) was fed to the induction plasma torch to determine the operating characteristics of the plasma torch from its ability to spheroidize alumina particles. The granular alumina was used as a stand-in for composite particles of uranium dioxide and graphite.

The independent variables investigated in these preliminary runs included alumina feed rate, power input to the oscillator, gas composition, and gas flow rate. The operating conditions and results for 21 runs with granular alumina are listed in Table III. Granular alumina was introduced from the top of the quartz tube into the plasma zone in all runs except Run IB-4, in which alumina was introduced 1 in. deeper into the plasma zone. Typical granular alumina and typical alumina after one pass through the induction plasma torch in Run IIC-3 are shown in Fig. 8.

Alumina feed rates between 0.6 and 1.2 kg/hr had little effect on the percentage of melted particles (e.g., compare the percent melted for Runs IIA-2 and IIA-3 or for Runs IIA-1 and IB-1), indicating that much higher feed rates may be practical and that the upper limit for feed rates of solids (where the solids start to cool the plasma flame) had not been reached. When power input to the oscillator was increased from 13.5 to 21 kW while the other operating conditions were maintained constant, the percentage of alumina particles melted increased from 13 in Run IIA-1 to 57 in Run IIA-2 and 49 in Run IIA-2R.

TABLE III. Operating Conditions and Results of Preliminary Plasma Torch Runs with Granular Alumina

Run	Power Input to Oscillator, kW	Alumina Feed Rate,[a] kg/hr	Feed Rate of Gas, scfh, through					Plasma Gas Composition, vol %			Reaction Chamber Pressure,[b] psig	Percent of Alumina Particles Melted
			Axial Nozzle	Tangential Nozzle B	Tangential Nozzle C	Radial Nozzle	Alumina Feeder	Argon	Nitrogen	Hydrogen		
IA-1	13.0	0.6		59		74	6.8	100			-6.4	21
IA-2	12.9	0.6		59		74	6.5	100			-5.4	3
IA-3	13.1	0.6		59		74	6.5	100			-5.4	3
IB-1	14.0	0.6		59		74	6.5	100			-5.6	16
IB-2	13.3	0.6		59		74	6.5	100			-5.6	15
IB-2R	13.3	0.6		59		74	6.5	100			-2.5	52
IB-3	14.0	0.6		59		74	6.5	100			-0.5	15
IB-4c	13.2	0.6		59		74	8.8	100			-5.6	1
IIA-1	13.5	1.26		59		74	6.5	100			-5.4	13
IIA-2	21.0	1.26		59		74	6.5	100			-5.6	57
IIA-2R	21.0	1.26		59		74	6.5	100			-5.6	49
IIA-3	21.0	0.6		59		74	6.5	100			-5.4	51
IIB-1d	13.0	0.6		59		74	6.5	100			-5.4	1
IIB-2d	21.0	0.6		59		74	6.5	100			-5.6	2
IIC-1	14.2	0.6		59		74	6.5	100			-6.4	21
IIC-2	15.2	0.6		59		74	6.5	96	4		-6.4	29
IIC-3	14.3	0.6		59		74	6.5	92	8		-6.4	89
III-1	13.0	0.6		59		93	6.5	96		4	-3.4	23
III-2	13.0	0.6	74	59			6.5	100			-4.9	42
III-3	13.0	0.6		59	74	74	6.5	100			-4.9	7
III-4	13.0	0.6	59			74	6.5	100			-4.9	27

[a]Duration of alumina feed for each run was 3 min.
[b]Reaction chamber contained air.
[c]The powder injection point was 1 in. below the normal location.
[d]Red granular alumina was fed in these two runs, whereas white granular alumina was fed in all other runs.
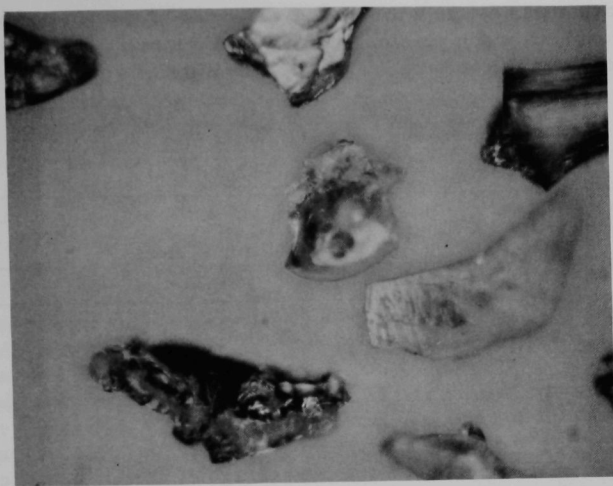
a. Granular Alumina Feed. Mag. 200X.
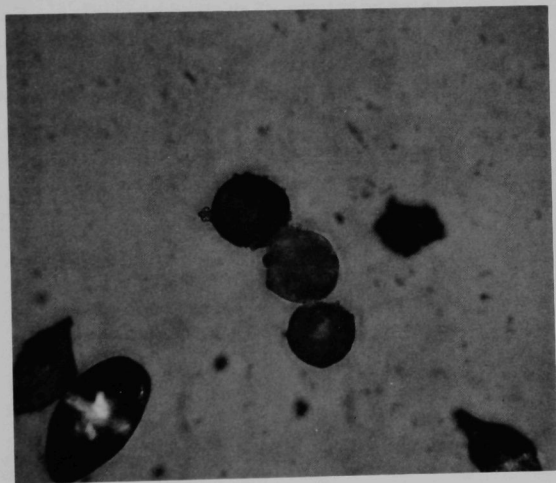
b. Spheroidized Alumina after One Pass through
the Induction Plasma Torch. Mag. 200X.

Fig. 8. Typical Granular and Spheroidized Alumina (Run IIC-3)

The effect of gas composition on spheroidization of alumina is shown in Table IV. The percentage of melted particles increased from 16 for pure argon plasma (Run IB-1) to 21 for 4 vol % nitrogen-argon plasma (Run IIC-1), and 89 for 4 vol % hydrogen-argon plasma (Run IIC-3).

Spheroidization was fairly uniform for all sizes of the particles (53-137 μm) with the hydrogen-argon plasma. With the other two types of plasma, a larger percentage of the smaller particles melted.

TABLE IV. Effect of Diatomic Gas in Argon Plasma
on Spheroidization of Granular Alumina

Solid feed rate: 10 g/min

Run	Diatomic Gas Concentration in Argon		Particles Melted, %					
	Gas	Vol %	137 μm	115 μm	96.5 μm	75 μm	53 μm	Total
IB-1	-	-	0	0	2	33	50	16
IIC-1	N$_2$	4	0	2	50	60	60	21
IIC-3	H$_2$	4	90	90	100	95	90	89

Increasing the plasma gas flow rate from 140 scfh in Run IA-3 to 159 scfh in Run III-1 increased the percentage of alumina particles melted from 3 to 23, but a further increase to 210 scfh in Run III-3 decreased the percentage melted to 7. Changing the gas flow patterns in the plasma had significant effects on the percentage of melted particles. For a constant gas flow rate, using the axial and tangential gas flow patterns in Run III-2 gave the highest percentage melted, 42, with the pure argon plasma flame at constant power input to oscillator, but the flame was unstable when there was any significant axial flow.

APPENDIX B

Runs with UO$_2$-Graphite Agglomerates

Agglomerated particles of UO$_2$ and graphite were fed into the tail flame of plasma in 17 runs. The operating conditions are listed in Table V, and the results of analyses of product samples are presented in Table VI.

During the first 11 runs, the reaction chamber contained air. Some graphite was burned. It was observed that the carbon content of the solid product decreased at higher power inputs (20 kW), lower gas flow rates (132 scfh), and higher hydrogen concentrations in the plasma gas (6 vol %). The effects of these operating conditions on lowering the carbon content of the agglomerates was analogous to their effects on the percentage of melted granular alumina particles. As was expected, the oxygen content of the product was nearly the same as the oxygen content of the feed, indicating that the hot particles emanating from the torch had reacted with atmospheric oxygen in the reaction chamber.

In the final set, six runs, uranium dioxide and graphite agglomerates were fed into the tail flame formed from argon-3.2 vol % hydrogen plasma in an inert-argon atmosphere. The results of the final set of runs were discussed in the main body of this report.

TABLE V. Operating Conditions for the Conversion of Uranium Dioxide-Graphite Agglomerates in the Induction Plasma Torch Reactor

Feed point for powder: Tail flame

Run	Power Input to Oscillator, kW	Duration, min	Powder Feed Concentration, wt % C	Powder Feed Concentration, wt % O	Average Particle Size, μm	Nominal Feed Rate, kg/hr	Feed Rate of Gas, scfh Through Tangential Nozzles	Feed Rate of Gas, scfh Through Radial Nozzles	Feed Rate of Gas, scfh Through Solid Feeder	Hydrogen in Plasma, vol %	Reaction Chamber Atmosphere	Reaction Chamber Pressure, psig
1	15.2	2	8.7	12.1	107	0.9	57	67	8	3.2	Air	-2.5 to 0
2	15.9	2	8.7	12.1	107	0.9	57	67	7	3.2	Air	-2.5 to 1
3	15.9	2	8.7	12.1	107	0.9	57	67	10	3.2	Air	0
4	15.7	2	8.7	12.1	107	0.6	57	67	8	3.2	Air	0
5	16.8	2	8.7	12.1	107	0.9	57	67	8	0.0	Air	-2.5 to 0
6	15.7	2	8.7	12.1	107	0.9	57	67	8	1.5	Air	0
7	15.0	2	8.7	12.1	107	0.9	55	67	8	6.0	Air	1.0
8	15.4	2	8.7	12.1	107	0.9	57	93	8	3.1	Air	0
9	20.0	2	8.7	12.1	107	0.9	57	93	8	3.0	Air	1.0
10	15.7	2	8.7	12.1	107	1.2	57	67	8	3.1	Air	-1.0
11	15.8	2	8.7	12.1	107	0.9	57	67	8	3.1	Air	1.0
12	15.9	2	8.7	12.1	107	0.9	57	67	7	3.2	Argon	0 to 2.5
14	15.9	2	8.7	12.1	107	0.9	57	67	7	3.2	Argon	-1.5 to 2.5
15	15.9	10	8.7	12.1	107	0.9	57	67	7	3.2	Argon	1.0
16	15.9	10	8.7	12.1	107	0.9	57	67	7	3.2	Argon	0.5 to 1.0
17	15.9	60	5.7	13.0	107	0.9	57	67	7	3.2	Argon	0.4 to 1.8
18	15.9	60	12.1	13.2	58	0.06	57	67	7	3.2	Argon	0.3 to 1.1

During Run No. 13, a porcelain insulator supporting a choke coil in the Lepel high-frequency generator broke, causing rf arcs. The run was prematurely terminated, and hence the operating conditions are not tabulated above.

TABLE VI. Analysis of Product Samples

Run	Carbon,[a] wt % Total[b]	Free	Oxygen,[a] wt %	X-ray Diffraction Analysis[c] UO$_2$	UC	Alpha-UC$_2$
1	6.3					
2	6.5		10.7			
3	6.2					
4	5.0					
5	5.5					
6	5.3		12.1			
7	5.0					
8	5.6					
9	4.9		12.4			
10	5.3					
11	5.8		10.5			
12				Major	Intermediate	Intermediate
14	6.2		9.6	Major	Intermediate	Intermediate
15	5.7	1.7	8.2	Major	Minor	Minor
16	5.7	1.7	7.9	Major	Intermediate	Intermediate
17	3.4	1.2	11.0	Major	Minor	Minor
18	8.4	3.6	7.4	Major	Intermediate	Intermediate

[a]Average of two analyses.

[b]Total carbon (determined by LECO combustion method) in the sample, including free carbon (determined by a gravimetric method, which includes dissolution of the sample in dilute nitric acid) and the carbon in UC and UC$_2$. The total carbon value for the sample is used in the analysis for oxygen content by combustion at 925°C of the sample.

[c]Estimated concentration ranges: Major = ~50 wt %; intermediate = 50 to 30 wt %; minor = 30 to 10 wt %. The compounds present are identified by the X-ray diffraction patterns generated by a sample of about 100 μg held in a capillary tube. The presence or absence of a compound is decided by visual inspection, and by comparison of the patterns generated with standard patterns of these compounds. Since the relative concentrations of these compounds are estimated by assessing the intensity of the patterns present, the values for the concentrations are only qualitative estimates.

APPENDIX C

Performance of Equipment Components

Equipment component performance pertinent to the further development of the induction plasma torch reactor for the conversion of uranium dioxide to uranium carbide and to the development of large-scale units is summarized in the following discussion.

1. Induction Plasma Torch Performance

During the preliminary series of runs, granular alumina was introduced from the top of the quartz tube directly into the plasma zone. The quartz tube and the hot gas exit nozzle at the bottom of the tube became coated with alumina particles in about 12-15 min of operation (four to five runs, each of 3-min duration). The presence of any foreign particles on the tube wall promotes arcing of the rf current to the tube, causing the tube to break. Hence, it was necessary to clean the tube after every fourth or fifth run.

In the first run to convert UO_2 to UC (not reported in Table V), agglomerates of UO_2 and graphite powder were fed directly (like the alumina) into the plasma arc of argon with 3 vol % hydrogen. The power input to the oscillator was relatively low (13 kW). The gas flow rates and flow patterns were such that a stable plasma was formed. However, soon after the flow of solids was started, the quartz tube cracked. A possible explanation is that this breakage might have been caused by localized overheating due to increase in heat transferred to the quartz tube when the agglomerates were present. The increase in the heat transfer may be attributable to the higher emissivity of the plasma when it contains the agglomerates than when it contains the granular alumina or no solids.

During the subsequent runs, the uranium dioxide-graphite agglomerates were fed into the tail flame downstream from the plasma zone (through orifices in the exit nozzle below the plasma-forming region). The absence of bulk of solids in the plasma zone allowed longer-duration runs to be made, although the solids were exposed to the high temperatures for shorter times than when they were fed directly into the plasma zone.

As the UO_2-graphite agglomerates were fed, a thin coating of fine powder formed on the inside of the brass double flange that holds the plasma torch and the quartz-plate insulator. Typical coating is shown in Fig. 9. The fines might have been generated by breakage of large agglomerates due to rapid evolution of CO product during conversion, and coating might have occurred due to electrostatic phenomena and/or thermal phenomena near the flange.

a. Torch Assembly before Run

Double Flange
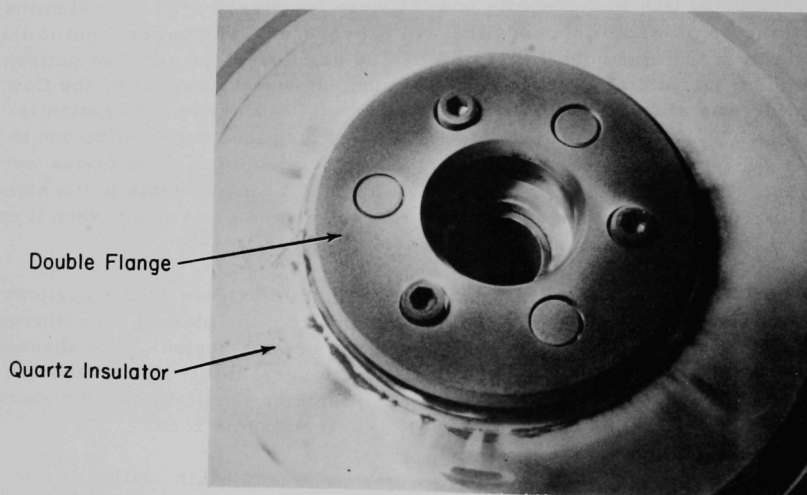
Quartz Insulator

b. Torch Assembly after Run

Fig. 9. Coating of Fines on Double Flange Holding Plasma Torch
and Quartz Plate Insulator during a Run

The performance of the induction plasma torch indicates that the designs of the plasma containment tube, the solids-feed orifices, and the auxiliary flanges should be improved for further development or scale-up of the conversion process.

2. Mullite Guide Tube

A mullite guide tube (5.6 in. ID, 6.1 in. OD, 27 in. long) was installed below the plasma torch to contain the tail flame and guide the particles into the product receiver. In the absence of the guide tube, powder fed into the plasma (both axially and radially) did not fall through the center of the tail flame. Instead, most of the powder was held up on horizontal ledges in the reaction chamber, and only a minor amount reached the product receiver.

The top of the mullite guide tube broke occasionally. Typical breakage of the top portion of the mullite tube is shown in Fig. 10. This breakage might have been caused by localized overheating of the guide tube.
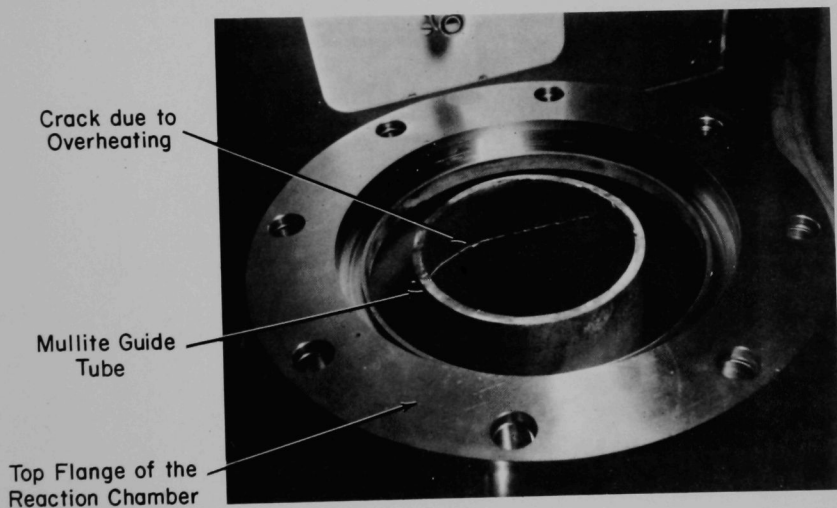

Fig. 10. Top Portion of Mullite Tube, Showing Breakage due to Overheating

To determine the approximate temperatures reached by the mullite tube during a run, marks were made longitudinally on outside wall of the tube with heat-sensitive pencils (Tempilstiks*) before a run. The tube was inspected after a run. The discolorations of the Tempilstik markings indicated that the top 18.5 in. of the guide tube had reached temperatures

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*Manufactured by Tempil Corporation, New York, N. Y.

greater than 500°F (255°C) and that the top 21 in. had reached temperatures greater than 300°F (149°C). An approximate value of the maximum temperature of the tube was calculated by assuming that the net heat gain (the difference between heat gained by radiation and forced convection from the plasma flame to the tube and heat lost by radiation and natural convection from the tube to the reaction chamber) resulted in a linear temperature profile along the length of the tube. The temperature gradient was calculated to be 80°F/in., and the highest temperature (at the top of the pipe) to be 1480°F (804°C). Further design would have to be done for a guide tube operated at these temperatures for further development or for scale-up of the conversion process.

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